MAGNETO-OPTICS OF ANTIFERROMAGNETS

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ABSTRACT

In some antiferromagnetic (AFM) crystals ions of different magnetic sublattices occupy crystallographic sites that are translationally nonequivalent and are not interrelated by a symmetry center. These crystals can have magneto-optic properties qualitatively different from those of other AFMs. In particular, one can observe birefringence and dichroism of linearly polarized light that are directly proportional to magnetic field strength. Both the linear magneto-optic effect (LMOE) and quadratic magnetic gyration (QMG) are sensitive to the crystal magnetic symmetry and to reorientation of magnetic sublattices of AFM crystal. The results of the symmetry analysis and experimental studies of LMOE and QMG in different AFM crystals are reported. The LMOE-induced changes of symmetry components of the dielectric tensor for AFM compounds containing Co\textsuperscript{2+}, Fe\textsuperscript{2+}, Mn\textsuperscript{2+} and Fe\textsuperscript{3+} ions proved to be sufficiently large. This makes it possible to visualize time-reversed AFM domains in the magnetic field of 10^3 - 10^4 Oe. The most pronounced experimental results were obtained for AFM CoF\textsubscript{2}.

INTRODUCTION

The lowering of the space-time symmetry of a medium as it becomes magnetically ordered leads to the appearance of physical properties which were not present earlier. Well known, for example, are piezomagnetism and the magneto-electrical effect, that have no analog among the properties of magnetically ordered substances. New spontaneous optical properties which appear on magnetic ordering are similar to the properties of magnetically disordered media magnetized by an external magnetic field. For example, the spontaneous rotation of the polarization plane in a ferromagnet is proportional to the magnetic moment, just as the field-induced Faraday rotation in a paramagnet is proportional to its magnetization in a magnetic field. An analogous correspondence can be seen between the spontaneous and field-induced effects of birefringence of linearly polarized light, as well as between the spontaneous and field-induced effects of gyrotropic linear birefringence. The multisublattice structure ferri- and antiferromagnets is no obstacle to such analogy. The optical properties of a multisublattice magnet can be represented as a simple superposition of the properties of the individual interpenetrating magnetized media.

However, optical properties of a magnetically ordered medium induced by the external actions can have no analog among induced optical properties of magnetically disordered media. The presence of magnetic subsystems in crystals may lead to induced optical effects forbidden in nonmagnetic crystals. Optical effects that are allowed only in magnetically ordered media are sensitive to the magnetic symmetry of the medium and to reversal of the directions of all the elementary magnetic moments of the media. These properties of new effects are attractive from the point of view of studying the symmetry of magnetically ordered substances and observation of time-reversed or collinear domains in compensated AFM.

In this article, we present the results of experimental studies and applications of two magneto-optic effects symmetry allowed in AFM crystals whose magnetic symmetry group does not involve the anti-inversion operation: LMOE and quadratic magnetic rotation. Both effects are forbidden in magnetically disordered substances.

SPACE-TIME SYMMETRY ANALYSIS

The optical properties of transparent crystals can be described by the dielectric constant tensor $\varepsilon_{ij}(\omega, \vec{r})$. The components $\varepsilon_{ij}$ are subject to the Onsager symmetry relations for the kinetic coefficients. For the time-invariant or magnetically disordered medium in a magnetic field these relations are of the simple form

$$\varepsilon_{ij}(-\vec{H}) = \varepsilon_{ji}(\vec{H})$$

(1)
They lead to even H-dependence of the tensor \( E_{ij} \) symmetric components describing birefringence of linearly polarized light, and odd H-dependence of antisymmetric components describing the Faraday rotation. But for the magnetically ordered media the time inversion operation \( \tau \) is not a symmetry operation and thus the Onsager relations (1) require generalization. The generalized symmetry relations for the kinetic coefficients involve space transformation operations \([1]\). For crystals described by Shubnikov groups with the account of the dependence \( E_{ij} \) not only on \( \omega \), but also on the wave vector \( k \) of the light wave, the generalized Onsager relations can be written in the most general form, viz.:

\[
E_{ij}(R \cdot H, R \cdot k) = R_{in} R_{jn} E_{mn}(H, k) \quad (2a)
\]

\[
E_{ij}(R \cdot H, R \cdot k) = R_{in} R_{jn} E_{mn}(H, k) \quad (2b)
\]

\[
E_{ij}(R \cdot H, R \cdot k) = R_{in} R_{jn} E_{mn}(H, k) \quad (2c)
\]

\[
E_{ij}(R \cdot H, R \cdot k) = R_{in} R_{jn} E_{mn}(H, k) \quad (2d)
\]

Here \( R \) are the operators of rotation of the symmetry point group, \( R = R \cdot \tau, \tau = R \cdot 1 \). The magnetic field strength \( H \) and vector \( k \) transform as:

\[
\begin{align*}
R \cdot H &= R \cdot H, \quad R \cdot H = -R \cdot H, \quad R \cdot H = -R \cdot H \\
R \cdot k &= -R \cdot k, \quad R \cdot k = -R \cdot k, \quad R \cdot k = R \cdot k
\end{align*}
\]

Confining ourselves to the second-order magneto-optic effects \( E_{ij} \) can be expanded in the components \( H \) and \( k \)

\[
E_{ij} = E_{ij,i} + E_{ij,i} H + E_{ij,i} k + E_{ij,i} H H + E_{ij,i} H_k + E_{ij,i} H \kappa + E_{ij,i} H \kappa + \sum E_{ij,i} H \kappa
\]

Here indices \( S \) and \( O \) denote symmetry and anti-symmetry of tensors with respect to the permutation of indices \( i \) and \( j \). It can be seen from Eq. (2b) and (2d) that symmetric and anti-symmetric tensors \( E_{ij,i} \) and \( E_{ij,i} \) vanish identically if a medium is magnetically disordered, or if it is ordered but its magnetic cell period is multiple of that of the chemical cell (operation 1 is a symmetry operation of a magnetic point group), or it has an anti-symmetry center (1-1 is a symmetry operation and 1 is a symmetry operation of the magnetic Laue group). But if a magnetic crystal is noncentroantisymmetric, \( E_{ij,i} \neq 0 \) and \( E_{ij,i} \neq 0 \) and in such a crystal birefringence of linearly polarized light, directly proportional to the field strength, is symmetry allowed, as well as magnetic rotation proportional to the square of a field. Moreover, in noncentrosymmetric magnetic crystals with a magnetic unit cell which is not multiple of a chemical one the H-field-induced natural magnetic rotation is possible (tensor \( L_{ij} = 0 \).

3. LINEAR MAGNETO-OPTIC EFFECT

Magneto-optic effect described by the axial c-tensor \( q_{ijs} \), symmetric in the first pair of indices, consists of inducing with a magnetic field linearly-polarized light birefringence directly proportional to the H-field. In contrast to the quadratic Cotton-Moutouen effect, it reverses sign when the field direction is reversed or the directions of all the elementary magnetic moments in the crystal are reversed. The fact that the magneto-optic effect studied is similar in its manifestation to the well known linear electro-optic effect (the Pockels effect) makes it possible to name it the Linear Magneto-Optic Effect (LMOE). Similar to the disappearance of the Pockels effect in centrosymmetric crystals LMOE is symmetry forbidden in centrosymmetric crystals. The matrix of the axial c-tensor \( q_{ijs} \) of a magnetic crystal is analogous to that of the polar \( l \)-tensor describing the linear electro-optic effect in a nonmagnetic crystal whose symmetry is defined by a set of operators of the magnetic crystal Laue group by replacing the proper antirotation operation with the improper rotations.

The symmetry of the magnetic ordering usually lends itself more easily to change by external action than the symmetry of the atomic ordering. In order to reveal the role of the ordering operation \( 1 \) is not as well as to estimate the temperature and field dependences of LMOE it is reasonable to express a change of components \( E_{ij} \) as a function of the parameters of the magnetic ordering. In this case it is necessary to regard the internal parameters which characterize the ordering as an external action on a crystal with a fixed symmetry of its ionic subsystem \([2]\). It is useful to choose the magnetic characteristics in the form of the magnetic vectors that are linear combinations of magnetic moments of the sublattices and transform in accord with the irreducible representations of the crystal space group \([3,4]\). For two-sublattice AFMs the magnetic vectors are the ferromagnetism vector \( M \) and antiferromagnetism vector \( L \). Matrices of the magneto-optic coefficients of the expression

\[
E_{ij} = E_{ij,i} + Q^{(LL)}_{ij,i} L_{\alpha} L_{\beta} + Q^{(MM)}_{ij,i} M_{\alpha} M_{\beta} + Q^{(ML)}_{ij,i} M_{\alpha} L_{\beta}
\]

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can be obtained by taking into account the transformation properties of the AFM and FM vectors. It is also necessary to take into account the fact that the effect of the crystal symmetry space operations can alter the internal magnetic property. For example, if the operation changes the numbering of the sublattices, then the direction of the AFM vector is changed. This transformation property of the magnetic vectors can be taken into account by introducing the even and odd space operations; and the following relations for the coefficients \( Q_{\gamma\phi}^{(ML)} \) can be written in the general form as

\[
Q_{\gamma\phi}^{(ML)} = \pm R_{\gamma\mu} R_{\phi\nu} R_{\mu\nu} Q_{\gamma\phi}^{(ML)}
\]

Here "\(+\)" and "\(-\)" correspond respectively to the even and odd operations of the space symmetry, \( R_{\gamma\mu} \) are the matrix elements of the operator \( R \) of the crystal Laue group. It can be seen that for a two-sublattice AFM the symmetry of the matrices \( Q_{\gamma\phi}^{(ML)} \) and \( Q_{\mu\nu}^{(ML)} \) in Eq.(5) is similar to the symmetry of the photoelastic tensor matrix. The symmetry of \( Q_{\gamma\phi}^{(ML)} \) can be obtained from Eq.(6). They are of the same symmetry as the corresponding matrices of the linear magnetostriction effect.

Let us consider the most illustrative experimental results, namely, the lowering of the crystal optical class of a collinear tetragonal AFM CoF\(_2\) in a longitudinal magnetic field \([5]\). Cobalt fluoride is a thoroughly investigated two-sublattice collinear AFM with clearly pronounced piezomagnetic properties. Its magnetic point group is \( 4/mmm \). Its magneto-optic tensor \( Q_{\gamma\phi} \) has the component \( q_{x\gamma} \) distinct from zero. For the linearly polarized light propagating in the \( C_4 \) direction the magnetic field \( H_{||C_4} \) should induce a birefringence equal to

\[
\Delta n = \frac{1}{2\alpha_0} Q_{x\gamma} H_{||C_4} = \frac{1}{2\alpha_0} Q_{x\gamma}^{(ML)} H_{||C_4} \quad (7)
\]

and the polarization directions of optical modes propagating in the crystal coincide with the axes \([110]_||x'\) and \([110]_||y'\). The sign of birefringence \((n_+ - n_-)\) is defined by the signs of the \( z \)-projections of the magnetic field and AFM vector. The lowering of the optical symmetry of this AFM is illustrated by the conoscopic figures of Fig.1 obtained from the sample which is in the same AFM state under the opposite orientations of a magnetic field. Figs.2 and 3 show respectively the magnitude of birefringence as a function of field strength and temperature. The effect disappears as the sample is heated above \( T_N \).

![Fig.1. Illustration of optical biaxiality induced by a magnetic field H||CoF2 in CoF2.](image)

As to the nature of the appearance of LMOE in CoF\(_2\), it should be noted first of all that, as the quadratic Cotton-Mouton effect, LMOE may be represented as a superposition of both a primary and secondary effects. The primary effect is due to a direct magnetic field influence on the energy structure of the crystal magnetic ion system, and the secondary one is connected with the deformation of inter-ion bonds and the crystal strains. Fig.2 shows the results of estimates of the secondary contribution obtained with the assumption of its photoelastic nature. It can be seen that magnetostriction strains of the CoF\(_2\) tetragonal structure are not basic.

The main contribution to LMOE seems to be due to the nonequivalent energy spectra of two collinear sublattices in a crystal. Co\(^2+\) ions belonging to different sublattices are translationally nonequivalent. An ion with a primary \( z \)-projection of the ions of a single sublattice has the low symmetry (the point group \( mmm \)) and possesses birefringent properties for linearly polarized light propagating along the \( C_4 \) axis. But the birefringence due to the ions of one sublattice is totally compensated by the ions of the other one. The magnetic field applied along the \( C_4 \) axis is parallel to the magnetic moment of the first sublattice and is antiparallel to the second. It violates this compensation since it makes the sublattices energy nonequivalent. The removal of degeneracy of the sublattice energy in the crystal spectrum can be one of the reasons for LMOE. The other reason can be related with quantum-mechanical state.

![Fig.2. Magnetic field dependence of linear birefringence for light propagating along the tetragonal axis in AFM CoF2. Two straight lines correspond to two time-reversed AFM domains. The dashed section is the contribution to the effect due to magnetostriction and photoelasticity of the crystal.](image)
mixing. This mixing results from the joint effect of a low-symmetry crystal field, spin-orbit interaction and magnetic field. It differs for Co\textsuperscript{2+} ions of two sublattices \cite{6,7}. A certain contribution can be due to the difference between populations of spin-wave subbands in a magnetic field when $T \neq 0$. This can lead to a change in the dispersion dependence of dichroism of the exciton-magnon absorption with the temperature increase.

Dichroism of linearly polarized light propagating along the tetragonal axis, odd in the magnetic field strength, is clearly observed in intra-configurational electronic transitions and particularly in the exciton-magnon absorption. Experiments on monodomain AFM samples of CoF\textsubscript{2} and MnF\textsubscript{2} showed that a magnetic field $H \parallel C_4$ induced the onset of linear dichroism in the most bands of the crystal absorption spectrum \cite{8}. Its magnitude increases with the increasing field, and the sign changes with the reversal of the $H$ field. With the fixed field direction the dichroism signs for two AFM states are different. Fig.4 illustrates the behaviour of magnetic dichroism in the exciton-magnon absorption band with the peak at 22 769 cm\textsuperscript{-1} in different experimental conditions. Characteristic dichroism S-like curves at low temperatures give evidence of the fact that the main contribution to linear dichroism, odd in a field, is due to the removal of sublattice degeneracy in the crystal energy spectrum. While approaching the Neel temperature the dichroism curve becomes of the $\Lambda$-like form, most likely, because of the decrease of the probability of optical excitation of a magnon in a sublattice whose magnetic moment is directed opposite to $H$ due to the thermal population of the excited spin states.

LMOE is also pronounced in CoCO\textsubscript{2}. A CoCO\textsubscript{2} crystal, uniaxial in the paramagnetic state becomes optically biaxial below $T_N$. While studying linear birefringence in the geometry $k \parallel C_4 || H$ it was found that the birefringence increases substantially with the field growth \cite{9}. At low temperatures birefringence is doubled in a field of about 75 kOe and in a field 300 kOe it is three times larger than the spontaneous one. At first glance an increase of birefringence in a field $H \parallel C_4$ is unexpected since the transverse projections of the magnetic sublattice moments can only decrease in a field. This increase is due to LMOE.

The number of AFM crystals revealing LMOE is not large. It was found in CoF\textsubscript{2}, at the perpendicular experimental geometry \cite{10}, in orthorhombic DyFeO\textsubscript{3} \cite{11}, tetragonal Ca\textsubscript{2}Mn\textsubscript{2}Ge\textsubscript{3}O\textsubscript{12} \cite{12} and trigonal Fe\textsubscript{5}O\textsubscript{6} \cite{13}. The field-induced birefringence of linearly polarized light was also observed in cubic Dy\textsubscript{2}Al\textsubscript{2}O\textsubscript{12}. It has different signs for the stable and metastable AFM states \cite{14} and, perhaps, due to LMOE.
4. MAGNETIC GYROTROPY QUADRATIC IN MAGNETIC FIELD STRENGTH

As mentioned above, the Onsager relations for the kinetic coefficients, generalized to magnetically ordered media, lift the prohibition against H-even terms in the expansion of the antisymmetric components of the dielectric tensor. The tensor describing the quadratic dependence of $\varepsilon_{ij}$ on $H$ identically is not zero in the crystals without the anti-inversion center. "Magnetic gyration, quadratic in $H$, can be also described by a third-rank axial $c$-tensor $G_{abc}$, dual to an antisymmetric with respect to the first two indices $c$-tensor $B_{abc}$. Unlike a symmetric with respect to two indices axial $c$-tensor $L_{a}$, which describes LMOE, the $G_{abc}$ tensor is symmetric with respect to the interchange of all three indices. The $G_{abc}$ tensor symmetry is the same as that of the $C_{a111}$ tensor describing quadratic in $H$ magnetization [15].

Quadratic in $H$ magnetic rotation of the polarization plane of light was observed in a collinear AFM CoF in the experimental geometry $k \parallel C_{4}$, $C_{4}H \parallel [110]$, [16]. Its value achieves 0.5 deg/mm under the applied field of 20 kOe for $\lambda = 6328 \AA$. The difference between quadratic and linear in $H$ magnetic gyration is distinctly observed in the absorption bands due to electronic transitions in the 3d-configuration of Co$^{2+}$-ions. The comparison of the magnetic dichroism spectra for $k \parallel C_{4}$, $H \parallel C_{4}$ and $k \parallel C$, $H \parallel [110]$ shows that dichroism can have different signs at the same signs of z-projections of the resulting magnetic moments, induced by fields $H \parallel C_{4}$ or $H \parallel [110]$, and for the retaining direction of the antiferromagnetic vector.

5. OPTICAL OBSERVATION OF TIME-REVERSED ANTIFERROMAGNETIC DOMAINS

The question of the possibility of experimental observation of collinear or 180-degree or time-inversed domains in compensated AFMs has been discussed many times. The first photographs of collinear AFM domains which are not disturbed by any external effects were obtained by polarized neutron topography [17]. The possibility of using optical methods for visualization of collinear AFM domains was considered in [18]. Time-reversed AFM domains were first observed optically in the complicated six-sublattice noncollinear Ising AFM Dy$_{3}$Al$_{12}$O$_{31}$ in a magnetic field near the metamagnetic phase transition [19]. The domains observed were not equivalent in energy differed slightly in their magnetic moments. Optical observation of time-reversed energy equivalent domains was carried out in a collinear AFM CoF by means of LMOE [20]. Birefringence of linearly polarized light due to longitudinal LMOE in CoF is accompanied by magnetic circular birefringence due to the Faraday effect. The transformation of polarization of light passing through the AFM$^+$ and AFM$^-$ domains in CoF at $H \parallel C_{4}$ are represented by means of the Poincare sphere representation in Fig. 5. One can see that a change in the $\lambda/4$-plate azimuth is necessary in dependence on magnetic field strength in order to receive the maximum contrast between domains. The photographs of Fig.6 show the AFM domains observed in CoF after thermocycling about $T_{N}$. The effect of quadratic magnetic rotation of the light polarization plane also made it possible to visualize time-reversed AFM domains. Thus, AFM domains were observed by using the quadratic magnetic rotation in CoF at a field of about 10 kOe.

The feasibility of visualization of AFM domains allowed to control the AFM...
FIG. 6. AFM time-reversed domain structures observed in CoFz at temperatures near T_n (3-5) K. There is no \( \lambda \frac{1}{4} \) plate in the observation set-up.

sample state while studying LMOE in CoFz, FeFz, DyFeO\(_3\), Ca\(_3\)Mn\(_2\)Ge\(_3\)O\(_{12}\), and QMG in CoFz, as well as to study processes of monodomainization and remagnetization of AFMs under the simultaneously applied magnetic field and elastic stresses and under the magnetic field deviation from the symmetry directions in CoFz. The possibility of experimentation on monodomain samples permitted to determine the magnetic point group of the complicated multisublattice noncollinear AFM \( \text{Ca}_3\text{Mn}_2\text{Ge}_3\text{O}_{12} \)\(^{[2]} \).

6. CONCLUSION

It can be readily seen that we have paid main attention to field-induced magneto-optic effects which have no analogs in magneto-optics of paramagnets. We considered only AFMs as new effects are the most prominent there. The values of birefringence due to both LMOE and QMG, as well as those of linear and circular dichroism, respectively, are sufficiently large and easily measurable. This permits us to apply those effects to visualization of time-reversed domains and determination of the magnetic point groups of AFMs. We also used these effects elsewhere for studying the magnetic phase transitions and peculiarities of energy spectra.

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